P-74

As-grown domain structure in lithium tantalate with inhomogeneous distribution of stoichiometry deviation

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We investigated the dependence of as-grown domain structure in $LiTaO_3$ (LT) on spatial distribution of Li concentration. Vapor transport equilibration process (VTE) was used for Li concentration increase from congruent (48.8 mol.%) to stoichiometric (50 mol.%) composition. Spatially inhomogeneous distribution was obtained by variation of VTE process time.

The Li concentration spatial distribution was measured by confocal Raman spectroscopy (Alpha AR300, Witech, Germany). Domain structure was visualized at the surface using optical (BX61, Olympus, Japan) and scanning electron (Merlin, Carl Zeiss, Germany) microscopies after selective etching and in the bulk using second harmonic generation microscopy (Ntegra Spectra, NT-MDT, Russia).

The inhomogeneous distribution was defined as the difference between surface and bulk Li concentrations (Δc). Three types of the distribution were defined: (a) near congruent composition with inhomogeneity near surfaces, (b) inhomogeneous distribution – stoichiometric composition at surfaces with decrease of Li concentration in the bulk, and (c) near stoichiometric composition.

The as-grown domain structure formed due to cooling below Curie temperature after VTE process. The formation of wide domain boundary due to phase transition depends on LT composition distribution. Congruent composition of LT led to formation of wide domain boundary consisted with maze domain structure. The narrow domain boundary was formed in LT with the inhomogeneous distribution due to toward moving of phase boundaries from surfaces to bulk. Domain structure in LT with near stoichiometric composition was represented by the interlaced layers of smooth head-to-head and zig-zag tail-to-tail charged domain walls.

The subsequent cooling to room temperature resulted in formation of isolated domains in the volume between surfaces and narrow domain boundary due to pyroelectric field. The isolated domains represented by both through and non-through domains with diameter up to 2 μm. The domain shape changed depending on local stoichiometry deviation from hexagonal for stoichiometric composition to trigonal for congruent one.

The different type of charged domain walls in ferroelectrics attracts interest due to possibilities of its applications.

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1. V.I. Pryakhina, E.D. Greshnyakov, B.I. Lisjikh, A.R. Akhmatkhanov, D.O. Alikin, V.Ya. Shur, A. Bartasyte, *Ferroelectrics* **525**, 47 (2018).